



Significant spatial gradients in new particle formation frequency in Greece during summer

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Abstract. Extensive continuous particle number size distribution measurements took place during two summers (2020 and 2021) at 11 sites in Greece for the investigation of the frequency and the spatial extent of new particle formation (NPF). The study area is characterized by high solar intensity and fast photochemistry and has moderate to low fine particulate matter levels during the summer. The average PM_{2.5} levels were relatively uniform across the examined sites. The NPF frequency during summer varied from close to zero in the south-western parts of Greece to more than 60 % in the northern, central, and eastern regions. The mean particle growth rate for each station varied between 3.4 and 8 nm h⁻¹, with an average rate of 5.7 nm h⁻¹. At most of the sites there was no statistical difference in the condensation sink between NPF event and non-event days, while lower relative humidity was observed during the events. The high-NPF-frequency sites in the north and northeast were in close proximity to both coal-fired power plants (high emissions of SO₂) and agricultural areas with some of the highest ammonia emissions in the country. The southern and western parts of Greece, where NPF was infrequent, were characterized by low ammonia emissions, while moderate levels of sulfuric acid were estimated (10⁷ molec. cm⁻³) in the west. Although the emissions of biogenic volatile organic compounds were higher in western and southern sectors, they did not appear to lead to enhanced frequency of NPF. The infrequent events at these sites occurred when the air masses had spent a few hours over areas with agricultural activities and thus elevated ammonia emissions. Air masses arriving at the sites directly from the sea were not connected with atmospheric NPF. These results support the hypothesis that ammonia and/or amines limit new particle formation in the study area.

1 Introduction

One of the key challenges in climate change is to quantify the exact role of atmospheric particles in the system. Atmospheric particles influence climate both directly, by absorbing or reflecting solar radiation, and indirectly, by becoming cloud condensation nuclei (CCN) and affecting the formation and characteristics of clouds (Seinfeld and Pandis, 2016). The aerosol net impact on the energy balance of the planet remains uncertain (IPCC, 2021). Although combustion and other sources can be a significant source of particle number in the atmosphere, new particle formation (NPF) is the dominant source of new particles in many environments worldwide (Kulmala et al., 2004; Kerminen et al., 2018). Atmospheric nucleation and subsequent growth of the formed fresh nuclei to larger sizes are an important, and sometimes the dominant, source of CCN on a global scale (Spracklen et al., 2006; Merikanto et al., 2009; Gordon et al., 2017). Also, since the formed particles belong in the ultrafine range (< 100 nm) they can have adverse health effects (Oberdörster et al., 2004; Politis et al., 2008).

The first step of NPF is the formation of a critical cluster through the synergistic activity of various gaseous precursors (Kulmala et al., 2001; Curtius, 2006). Although several compounds have been shown to participate in atmospheric nucleation, large uncertainty and often conflicting results exist regarding the mechanisms that drive the phenomenon in different areas. Sulfuric acid is the key component of the new particles in most environments (Kulmala and Laaksonen, 1990; Erupe et al., 2010; Lee et al., 2019). The binary sulfuric acid–water nucleation was the one of the first mechanisms studied (Kulmala and Laaksonen, 1990), but it does not explain the high nucleation rates observed in the troposphere (Weber et al., 1999). Both laboratory experiments and ambient measurements have provided evidence that ammonia, amines, highly oxidized molecules, and other organic and inorganic compounds can be involved in nucleation (Ripinen et al., 2011; Kirkby et al., 2011, 2016; Almeida et al., 2013; Lehtipalo et al., 2018). Despite this progress, the dominating new particle formation mechanisms in most areas remain uncertain.

Quantifying the spatial extent of NPF events is important for understanding the impact of the formed particles on climate. However, the available direct evidence is limited because simultaneous measurements at multiple locations are required. Regional NPF events, taking place on spatial scales exceeding 100 km have been reported (Kerminen et al., 2018). Bousiotis et al. (2021) analyzed measurements from 13 sites in Europe and found that regional events (occurring at both urban and rural sites) account for 5 % of the events in Spain and up to 60 % of the events in Germany. Berland et al. (2017) characterized the NPF occurrence and characteristics on three Mediterranean islands (Mallorca, Corsica, Crete) and found that on 8 of the 41 d of the campaign, NPF occurred at at least two of the three

sites. It is not clear if the events also covered the rather large marine areas between these islands. Further to the east, Baalbaki et al. (2021) found significantly higher NPF frequencies in Cyprus compared to the closest station, Finokalia, Crete, about 670 km away (Pikridas et al., 2012). In another study in the Eastern Mediterranean by Kalkavouras et al. (2021), 6 months of measurements in Athens, Finokalia, and Amman (Jordan) were analyzed. Only five concurrent NPF episodes were found at all three sites, all connected with northern air masses. Meanwhile, local NPF and growth (scales of tens of km) can take place inside plumes of combustion sources at coastal sites or in urban areas (Kerminen et al., 2018). Kammer et al. (2023) estimated that the NPF spatial scale at an agricultural site in France is 20 km or a little more.

The few previous relevant studies in Greece suggest significant spatial variability in the NPF frequency during summertime. Patoulias et al. (2018) reported that during June 2012 the measured NPF frequency at Eptapyrgio, Thessaloniki, in northern Greece, was close to 80 %. However, measurements between June and October 2009 at the same location showed an event frequency close to 30 % (Siakavaras et al., 2016). Athens is characterized by an intermediate to low NPF frequency, varying from 10 % to 30 % during summertime months (Petäjä et al., 2007; Vratolis et al., 2019; Kalkavouras et al., 2020). In Patras, in the northwest Peloponnese, although NPF is infrequent (about 10 %) (Patoulias et al., 2018; Aktypis et al., 2023), during 30 % of the days new particles appear at the site, which had been formed upwind 100–150 km northeast of the city (Aktypis et al., 2023). At the Navarino Environmental Observatory (NEO), in the southwest Peloponnese, a low frequency of around 5 % during the summer was found by Hansson et al. (2021). Furthermore, on the island of Crete, Kopanakis et al. (2013) reported only two NPF events in 54 summer days (3 %) in Chania, which is situated in the northwestern part of the island. Several studies at the Finokalia station, also in Crete but in the northeastern part of the island, suggest that the NPF frequency varies from 10 % to 30 % during summertime (Pikridas et al., 2012; Kalivitis et al., 2019; Kalkavouras et al., 2020). The spatially variable NPF in Greece during the warm period suggested by the above studies makes the country an excellent natural laboratory to study factors that may be limiting or enhancing the phenomenon in different areas. Pikridas et al. (2012) found that in Finokalia, NPF events occurred only when particles were neutral, while the non-events were related to more acidic particles. A base, probably ammonia, appears to control NPF in that region (Pikridas et al., 2012). The role of ammonia in NPF was also investigated in Patras (where the events are infrequent) using a dual-chamber system (Jorga et al., 2023). After filling both chambers with ambient air, ammonia was added into one of the two chambers of the system. The addition of ammonia resulted in NPF in the perturbed chamber in three-quarters of the conducted experiments, indicating that it could indeed be the missing ingredient for

atmospheric NPF in Patras during summertime (Jorga et al., 2023).

In this study we quantify the NPF frequency during two consecutive summers (2020 and 2021) at 11 different locations in Greece. This represents a high spatial density of measurements, considering the small size of the country (scale around 800 km). We focus on the summer periods because conditions favorable for NPF (high solar radiation, lower condensation sinks, higher biogenic volatile organic compound emissions; Nieminen et al., 2014; Bousiotis et al., 2021) are present. This makes summertime an excellent period to investigate factors limiting new particle formation because a high NPF frequency is expected everywhere. We take advantage of the significant spatial gradients in the frequency, behavior, and characteristics of the events in an effort to gain insights into the dominant new particle formation mechanisms in the Eastern Mediterranean and other similar regions.

2 Materials and methods

Continuous particle number size distribution measurements took place during two summers (2020 and 2021) as a part of the PANACEA project at 11 different locations in Greece. This coordinated effort included measurements in six of the largest Greek cities (Athens, Thessaloniki, Patras, Xanthi, Ioannina, Chania); a coastal site near a small village, Methoni (only in 2020); three remote areas (Finokalia, Lesvos (only in 2021), and Sifnos); and one elevated remote site (Mt. Helmos). The measurements took place between 1 August and 30 September during the 2020 summer campaign and between 15 July and 15 September during the 2021 summer campaign.

2.1 Sampling sites

The measurement sites were chosen to cover most of the country (Fig. 1) and to also represent different types of environments. The sites were classified as urban (URB), suburban (SUB), rural (RUR), and remote (REM) according to the criteria proposed by Larssen et al. (1999). In addition, they were also further characterized as coastal, continental, or high-elevation sites depending on their location. Detailed descriptions of the sampling sites are provided below.

2.1.1 Athens (ATH^{SUB})

Athens is the capital and largest city of Greece, with a population of approximately 4 000 000 inhabitants. The city is densely populated and is surrounded by mountains in the western, northern, and eastern sectors, facing the Saronic Gulf in the south. The measurements in Athens were performed at the National Centre for Scientific Research “Demokritos” (37°59′42″ N, 23°48′57″ E at 270 m a.s.l.) a suburban background site located about 8 km northeast of

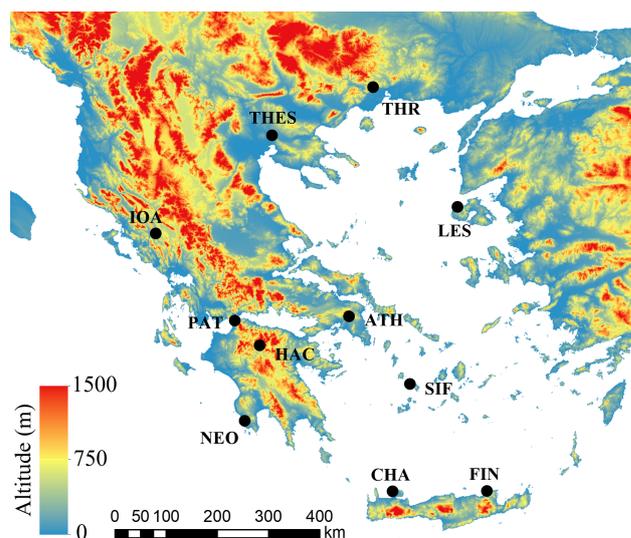


Figure 1. Elevation map of Greece indicating the 11 measurement sites (map created using the European Digital Elevation Model (EU-DEM) data, by Copernicus–EEA for eastern Europe).

the city center. The station is located in the foothills of Mt. Hymettus in a pine forest area and is affected by transported pollution from the city, especially during daytime hours (Eleftheriadis et al., 2021). The station can also be influenced by traffic emissions from a highway 400 m to the southeast and a major road 1.5 km to the north.

2.1.2 Thessaloniki (THES^{URB})

Thessaloniki is the second-largest city of Greece, with a population of approximately 1 100 000 inhabitants. The city faces the Thermaic Gulf in the southwest, while it is surrounded by Mt. Hortiatis in the east and the north. The western part of Thessaloniki is flat, and one of the largest agricultural areas of Greece begins 15 km from the city center. The measurements were conducted at the urban monitoring station of the Aristotle University of Thessaloniki (40°38′15″ N, 22°56′30″ E at 43 m a.s.l.) located in the city center. Traffic emissions from a nearby major road (Eleftheriou Venizelou Street) influence the site (Siakavaras et al., 2016). Other notable anthropogenic activities include an industrial zone 15 km to the northwest and a cement plant 7 km north of the sampling site. More information about the site can be found on Siakavaras et al. (2016) and Samara et al. (2014).

2.1.3 Patras (PAT^{SUB})

Patras is the third-largest city of Greece, with a population close to 300 000 inhabitants, and is located in the foothills of Mt. Panachaiko facing the Gulf of Patras. The measurements were conducted at the Institute of Chemical Engineering Sciences (ICE-HT). This is a suburban background site located approximately 8 km NE of the city center of Patras

(38°17′54″ N, 21°48′34″ E at 100 m a.s.l.). The site is surrounded by low vegetation and olive trees, while notable nearby human activities include the Patras–Athens highway (1 km away), a cement plant (6 km away), and the port of Patras (11 km away).

2.1.4 Ioannina (IOA^{SUB})

Ioannina is a city in northwestern Greece, with a population of 110 000 inhabitants. The city is surrounded by mountains, forming a plateau, and lies on the western shore of Lake Pamvotis. The suburban sampling site of Ioannina was at the University of Ioannina (39°36′55″ N, 20°50′12″ E at 500 m a.s.l.), located 6 km south of the city center. The area is characterized by low vegetation, and the major road leading to the city center is 2 km east of the sampling site. Due to the combination of its basin-like characteristics and the existence of a lake next to it, the city has a unique microclimate. It is characterized by calm winds and stable conditions and high relative humidity often resulting in fog events (Pilidis et al., 2005; Houssos et al., 2012).

2.1.5 Thrace (THR^{RUR})

Xanthi is a city in northeastern Greece with a population of around 70 000. The city lies in the foothills of the Rhodope Mountains (to the north and northwest), while the land is mostly flat in the other directions. The Aegean Sea is 20 km to the south of the city. Measurements were conducted in Xanthi only during the summer of 2020 in the rural–peri-urban site in the Democritus University of Thrace (41°09′00″ N, 24°55′12″ E at 75 m a.s.l.), located 4 km east of the city center. The nearby area is characterized by low vegetation, while a major road is located 500 m to the south. The measurements in Thrace during the summer of 2021 were conducted in Xylagani, a village with a population of 1200 inhabitants located about 45 km to the southeast of Xanthi. For this study we implicitly assume that the NPF events observed at both locations were representative of the broader Thrace region and do not differ significantly from each other. Although the two sites can have significant differences regarding local ultrafine particle emissions, the present study focuses on NPF events (lasting for > 2 h) that take place on a spatial scale larger than the distance (45 km) between them. In addition, the fine PM concentration levels of the two sites were compared and were found to be similar. The HYSPLIT analysis revealed that both areas are generally affected by the same air masses (mostly north and northeast), which is important when studying NPF.

2.1.6 Lesvos (LES^{REM})

Lesvos is an island in the northeastern Aegean Sea, located 10 km from the western coast of Turkey. It is the third-largest Greek island with a population of about 110 000 inhabitants.

The measurements during the summer 2021 campaign were performed at a remote site 7 km from the village of Sigrí (39°13′52″ N, 25°56′08″ E at 636 a.s.l.), in the western part of the island. The village has a population of 300 inhabitants. The nearby area is characterized by low vegetation and a small road 200 m to the north. Lesvos is one of the sites that is greatly affected by the Etesians, which dominate Greece and especially the Aegean Sea during the summer (Tyrlis and Lelieveld, 2013; Dafka et al., 2016).

2.1.7 Sifnos (SIF^{REM})

Sifnos is a small island in the southwestern Aegean, with a population of approximately 3000 inhabitants. The measurement site was located on the eastern outskirts of the village of Exampela (36°58′03″ N, 24°43′48″ E at 220 m a.s.l.) where no cars are allowed. The nearest local traffic road is 250 m to the west. The site is characterized as remote and is directly exposed to the northern flow of the Etesians, which dominate during the summer.

2.1.8 Finokalia (FIN^{REM})

Finokalia is a small rural settlement (< 10 inhabitants) located on the northeastern coast of the island of Crete. The atmospheric observation station of the University of Crete is located 3 km north of the village (35°20′15″ N, 25°40′11″ E at 250 m a.s.l.), facing the Aegean Sea. It is characterized as a regional background station and is representative of the Eastern Mediterranean. There are no notable anthropogenic activities (Mihalopoulos et al., 1997) within 15 km. The city of Heraklion (200 000 inhabitants) is located 50 km to the west.

2.1.9 Chania (CHA^{URB})

Chania is a coastal city in the northwestern part of Crete, with a population of 60 000. The measurements in Chania were conducted at the urban background station of the Technical University of Crete in Akrotiri (35°31′48″ N, 24°03′36″ E at 137 m a.s.l.). The station is located 5 km northeast of the city center and 2 km downwind of the sea. The nearby area is characterized by low vegetation, residential areas, and farms. A more detailed description of the Akrotiri station can be found in Lazaridis et al. (2008).

2.1.10 Methoni (NEO^{RUR})

The measurements (only for the 2020 summer campaign) were conducted at the Navarino Environmental Observatory (NEO), a rural site located in the southwestern part of the Peloponnese (36°49′20″ N, 21°42′14″ E at 30 a.s.l.). The site is located at Methoni, a village with 2600 inhabitants. Since the site is situated 400 m from the sea, it is also considered a coastal site. Low vegetation and olive trees characterize the nearby area.

2.1.11 Mt. Helmos (HAC^{REM})

The Helmos mountain Hellenic Atmospheric Aerosol and Climate Change station (HAC)² is a high-altitude station in Greece, operated by the Environmental Radioactivity Laboratory of National Centre for Scientific Research (NCSR) “Demokritos”. The station is located at the peak of the Helmos mountain in the northern Peloponnese (37°59′02″ N, 22°11′45″ E at 2314 m.a.s.l.). It is the only Greek station characterized as a free-tropospheric background site (Colaud Coen et al., 2018).

2.2 Instrumentation

Scanning or differential mobility particle sizers (SMPSs or DMPSs) were used for the measurements at the various sites. At least one SMPS or DMPS system was deployed at each of the stations for the measurement of the particle number size distributions. Different models of classifiers, differential mobility analyzers (DMAs), and condensation particle counters (CPCs) were used at each site (Table 1) covering at least the diameter range from 14 to 430 nm. During the summer 2021 campaign in Patras, the use of a second SMPS (classifier model 3080, CPC model 3775, DMA model 3085, TSI) together with a particle size magnifier (PSM), allowed the measurement of the aerosol concentrations and size distribution down to 1.3 nm.

Supporting measurements of black carbon (Aethalometer model AE33 and/or multi-angle absorption photometer (MAAP)), NO_x, and/or SO₂ were also performed at several stations (Table 1) and were used for the interpretation of the origin of the nucleation mode particles especially at urban and suburban stations like Athens and Patras. Measurements of several meteorological parameters were also performed during the two campaigns and included temperature, relative humidity, wind direction and speed, solar radiation, and precipitation. The meteorological stations were located either on site or a few kilometers away from the corresponding measurement site. For Thessaloniki and Thrace, the meteorological data were obtained from the National Observatory of Athens network that is described by Lagouvardos et al. (2017).

3 Data analysis

3.1 Event classification

The identification and classification of NPF events was performed according to the method proposed by Dal Maso et al. (2005). The whole available size range at each site was utilized for the classification of the days. Excluding Patras, the lowest detectable size at all stations varied between 9 and 14 nm, which (although it introduces some uncertainty) makes the size distributions comparable for the classification step. In Patras, the classification was performed twice: (a) by

accounting for both the full-size distributions and (b) accounting for only the ones for the size range 14–700 nm. The number of NPF events in Patras was the same with both methods. Information about the sub-10 nm particles in Patras was valuable for the identification and interpretation of the undefined events, observed frequently in that area (Aktypis et al., 2023). The main criteria for an episode to be classified as NPF include (a) the appearance of a distinct particle mode with a diameter below 25 nm (nucleation mode), (b) its persistence for at least 1 h, and (c) its subsequent gradual growth towards larger sizes. NPF events were further classified as (a) Class I if during the event the evolution of particle concentration and size could be observed clearly and the dynamic properties (i.e., particle formation and growth rates) could be estimated with a high confidence level and (b) Class II if fluctuations in nucleation mode particle concentration and/or diameter were observed. Weak (events when new particles grew in size, but either their number concentration was quite low or their growth was limited) or short-lived events (but still lasting for > 1 h) were also classified as Class II in this study. Days that did not meet the above criteria but during which the appearance of nucleation mode particles was evident were classified as undefined. These cases included days where either the growth of the observed particles was insufficient or their initial formation was not observed, even if they exhibited some growth.

Supporting measurements of BC, NO_x, and/or SO₂ were inspected (when available) to help identify the origin of the nucleation mode particles observed during a potential NPF event because primary particulate matter (PM) emissions can often cause biases in the classification process, especially in urban and suburban areas. Days when nucleation mode particles were observed at the same time with significant increases in concentrations of black carbon or NO_x or SO₂ were classified as non-events (although a few of them could be events, affected by local anthropogenic sources and photochemistry; Dai et al., 2017). However, if during an event, several concentration peaks from nearby sources obscured the NPF event but the formation and growth of new particles was still evident (Fig. S1 in the Supplement), the day was classified as an NPF event (Kulmala et al., 2012). Although a great effort was made to accurately classify all days, due to the subjectivity involved in the method, small uncertainties in the final results are expected.

3.2 Air mass origin

The origin of the air masses was investigated calculating the 24 h backward air trajectories that arrived at each measurement station during the two summer campaigns. One trajectory was estimated for each hour (24 trajectories per day). The calculation was performed using the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPPLIT-4) model developed by the NOAA Air Resource Laboratory (Draxler and Hess, 1998) in the backward trajectory

Table 1. Instrumentation, size range, and temporal resolution for each measurement site during the 2020 and 2021 summer campaigns.

Observable	Instrument/model	Size range (nm)	Resolution	Period
Athens (ATH–Suburban)				
Size distribution	SMPS (Classifier 3080, DMA 3081, CPC 3022A, TSI)	10–550	5 min	2020 and 2021
Black carbon	Aethalometer AE33, Magee Scientific		1 min	2020 and 2021
Thessaloniki (THES–Urban)				
Size distribution	SMPS (model 3034, TSI)	10–487	5 min	2020 and 2021
Patras (PAT–Suburban)				
Size distribution	SMPS (Classifier 3080, DMA 3081, CPC 3787, TSI)	14–750	4 min	2020 and 2021
	SMPS (Classifier 3080, DMA 3085, CPC 3775, TSI)	4–65	4 min	2021
	PSM (A11 nCNC, Airmodus)	1.3–4	4 min	2021
NO _x , NH ₃	NO _x and NH ₃ analyzer T201, Teledyne		1 min	2020 and 2021
SO ₂	SO ₂ analyzer 43i-TLE, Thermo Scientific		1 min	2020 and 2021
CO	CO analyzer API 300E, Teledyne		1 min	2020 and 2021
Black carbon	Aethalometer AE33, Magee Scientific		1 min	2020 and 2021
Ioannina (IOA–Suburban)				
Size distribution	SMPS (Classifier 3080, DMA 3081, CPC 3775, TSI)	14–723	3 min	2020 and 2021
Thrace (THR–Rural)				
Size distribution	SMPS (Classifier 3080, DMA 3081, CPC 3787, TSI)	14–723	3 min	2020 and 2021
Lesvos (LES–Remote)				
Size distribution	SMPS (custom built, CPC 3786, TSI)	9–430	5 min	2021
Sifnos (SIF–Remote)				
Size distribution	SMPS (model 3034, TSI)	10–487	5 min	2020 and 2021
Black carbon	Aethalometer AE33, Magee Scientific		1 min	2020 and 2021
Finokalia (FIN–Remote)				
Size distribution	SMPS (custom-built TROPOS type, CPC 3772, TSI)	9–850	5 min	2020 and 2021
Black carbon	Aethalometer AE33, Magee Scientific		1 min	2020 and 2021
Chania (CHA–Urban)				
Size distribution	SMPS (Classifier 3082, DMA 3081, CPC 3775, TSI)	12–593	5 min	2020 and 2021
Methoni (NEO–Rural)				
Size distribution	DMPS (custom built, CPC 3772, TSI)	10–806	15 min	2020
Mt. Helmos (HAC–Remote)				
Size distribution	SMPS (custom built TROPOS type, CPC 3772, TSI)	10–800	5 min	2020 and 2021

mode (Stein et al., 2015). A default arrival height equal to 50 m a.g.l. was chosen. For both summers, the NOAA Global Forecast System (GFS 0.25°, NOAA) meteorological files were used. The Weather Research and Forecasting (WRF) model (Skamarock et al., 2008) was also used for the calculation of the meteorological fields for the summer of 2021, providing a more detailed perspective of the air mass circulation over Greece. WRF was used with a 4 km grid resolution for Greece that was nested into a 12 km grid for the rest of Europe. The calculated trajectory endpoints were further

analyzed using the Openair package (Carslaw and Ropkins, 2012).

3.3 Calculation of the condensation and coagulation sinks and the growth rate

The condensation sink (CS) is an important parameter, affecting the rate with which vapors can condense on pre-existing atmospheric aerosol (Kulmala et al., 2001). It can

be calculated according to

$$CS = 2\pi D \sum_i \beta_i d_{pi} N_i, \quad (1)$$

where D is the diffusion coefficient of the condensable vapors, β_i the transition regime correction factor (Sutugin and Fuchs, 1970), d_{pi} the diameter of the SMPS size channel i , and N_i is the number concentration of that size channel. The diffusion coefficient of H_2SO_4 and a mean free path equal to 123 nm were chosen for the calculations in this work (Erupe et al., 2010; Dinoi et al., 2021).

The growth rate (GR) of the newly formed particles during an NPF event was calculated following the method described by Kulmala et al. (2012). Initially, the log-normal particle number size distributions were fitted using a least-squares fitting algorithm, and the geometric mean diameters of the nucleation, Aitken, and accumulation mode particles were determined. Next, the growth rate was determined as the slope between the starting time and diameter (D_{p1}) of an event and the time when the particles reached the 25 nm mobility diameter limit (D_{p2}), according to

$$GR = \frac{\Delta(25 \text{ nm} - D_{p1})}{\Delta t}, \quad (2)$$

where Δt is the time needed for particles with diameter D_{p1} to reach 25 nm. Because the smallest detectable particle diameters vary from site to site (Table 1), uncertainties are introduced in the direct comparison of the calculated growth rates. For the calculation of the growth rates, only the Class I events were considered because the evolution of the geometric mean diameter could be followed with a good confidence level. The starting time of each event was defined as the time when the observed nucleation mode geometric mean diameter started increasing.

4 Results and discussion

4.1 Ambient conditions during the measurements

Summer in Greece is characterized by relatively high temperature and sunlight intensity and relatively dry conditions. The temperature across all stations varied between 11 and 39 °C during the summer 2020 campaign, having an average of 25.8 ± 3.3 °C and between 16 and 41 °C during the summer 2021 campaign with an average of 26.8 ± 4 °C. The average temperature during the expected NPF occurrence periods (09:00–15:00 LT) was 28.5 °C. The summer 2021 campaign was characterized by some periods with extremely high temperatures due to a heat wave that peaked at the beginning of August (Founda et al., 2022). The average relative humidity of all stations was 60.1 ± 14.4 % and 57.7 ± 14.8 % for the two campaigns, respectively. The peak of the solar radiation exceeded 800 W m^{-2} around noon during most days of the campaigns, while rain events were overall rare (less than five, mostly occurring at the end of September). Average meteorological parameters for the two measurement periods and

each site can be found in Table S1 for 2020 and Table S2 for 2021.

The atmosphere was relatively clean in Greece during the measurement periods. Dimitriou et al. (2023) analyzed measurements of $PM_{2.5}$ from 14 PurpleAir sensors in Athens, Thessaloniki, Patras, Xanthi, and Ioannina between July 2019 and June 2021 and found that the average $PM_{2.5}$ levels were low (with a small variation from 8 to $10.8 \mu\text{g m}^{-3}$) during the summer. Even more interesting was the relatively homogeneous distribution of $PM_{2.5}$ across the country. This suggests that, with the exception of the free-tropospheric station of Helmos, the other 10 stations had similar $PM_{2.5}$ and sunlight levels.

The average N_{14} (mean number of particles with diameters larger than 14 nm calculated for both campaigns) concentrations varied significantly (Fig. 2). Helmos^{REM}, Chania^{URB}, Sifnos^{REM}, Finokalia^{REM}, and NEO^{RUR} had the lowest average N_{14} ranging from 2000 to 3000 cm^{-3} . Somewhat higher number concentrations were observed in Patras^{SUB}, Lesvos^{REM}, and Athens^{SUB}, where the average N_{14} varied between 3000 and 4000 cm^{-3} . Athens had higher than average nucleation mode particles mainly due to primary emissions (Eleftheriadis et al., 2021). Thrace^{RUR} had an average N_{14} of about $4170 \pm 3200 \text{ cm}^{-3}$, while in Ioannina^{SUB} the N_{14} was $6910 \pm 3200 \text{ cm}^{-3}$. Finally, Thessaloniki^{URB} had by far the highest average N_{14} compared to the other sites, equal to $10380 \pm 5900 \text{ cm}^{-3}$. The high particle concentration observed in Thessaloniki can be explained by the urban characteristics of the measuring station and by the frequent and intense NPF events identified at that site. Except for Ioannina, the average N_{14} of the two campaigns at each station was quite similar.

The nucleation mode (N_{14-25}) particles accounted on average for 12 ± 9 % (the latter number represents 1 standard deviation from the mean) of the total particle number concentration (N_{14}), with the corresponding fraction varying from 3 % in Chania to 35 % in Thessaloniki. The Aitken mode (N_{25-100}) particles accounted on average for 54 ± 5 % of N_{14} varying from 47 % in Thessaloniki to 62 % in Lesvos. Finally, the accumulation mode (N_{100}) particles corresponded to 34 ± 8 % of N_{14} varying from 18 % in Thessaloniki to 45 % in Chania. The N_{14} and N_{100} were calculated accounting for the upper size limit available at each site. This ranged between 430 and 800 nm, but the number contribution of these particles to the total ($N_{430-800}/N_{14}$) was negligible (always below 2 %) compared to that of the smaller particles.

4.2 NPF frequency

A total of 941 daily particle number size distribution plots (average data coverage was 76 %) were visually inspected together with the corresponding evolution of the nucleation, Aitken, and accumulation mode number concentrations in order to be classified as Class I NPF events, Class II NPF events, or undefined or non-events. Typical examples of

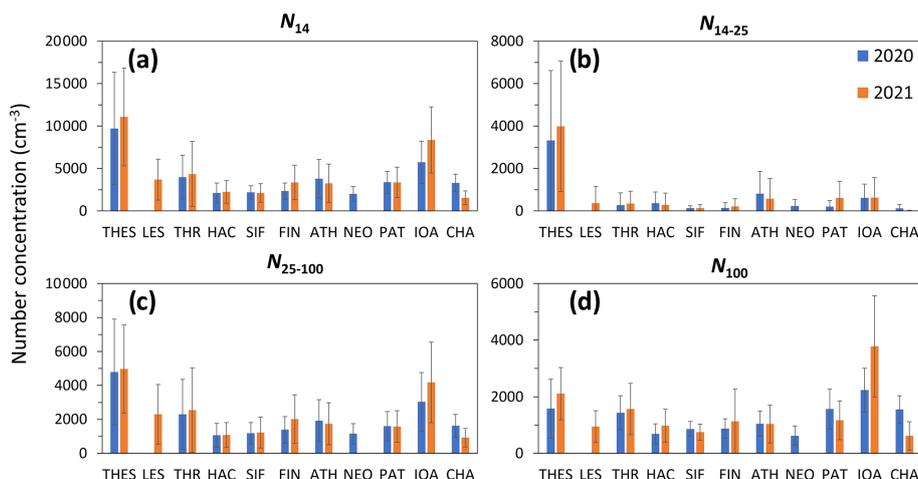


Figure 2. The average particle number concentrations of (a) the N_{14} , (b) the nucleation mode (14–25 nm), (c) the Aitken mode (25–100 nm), and (d) the accumulation mode (> 100 nm) at each measurement site during the summer campaigns of 2020 and 2021. The error bars represent 1 standard deviation.

each different category together with their corresponding N_{14} number concentration are depicted in Fig. 3. The formation and growth of new particles is clear in the Class I NPF event in Sifnos on 14 August 2020 (Fig. 3a). Newly formed particles appeared at around 10:00 LT and grew for 6 h reaching sizes of 100 nm. During the Class II event (Fig. 3b) in Finokalia on 23 July 2021, new particles were observed at 08:00 LT, but the evolution of their diameter and number concentration was not continuous. In the undefined event shown in Fig. 3c in Patras on 27 July 2021, new particles with a mean diameter of 25 nm appeared at around 11:00 LT, but their formation and their growth were absent. This day was classified as a transported event by Aktypis et al. (2023). Finally, Fig. 3d shows a day during which no NPF was observed in Chania.

A significant spatial variability in the NPF frequency (calculated as the total number of Class I and II events divided by the total number of days with available data) was observed across the 11 different areas during both summer campaigns (Fig. 4). The average NPF frequency varied from close to zero in NEO and Chania to 64 % in Thessaloniki. Although a substantial variation in the frequency at a given station from year to year is possible (Dal Maso et al., 2005; Manninen et al., 2010; Kalivitis et al., 2019), no significant differences (less than 10 %) were observed in the Greek stations between the two campaigns, except Finokalia.

The three stations (THES^{URB}, LES^{REM}, THR^{RUR}) in the north and the northeast all had intense events and high NPF frequencies (defined here as more than 40 %). Thessaloniki had the highest NPF frequency (64 %) compared to the other sites. The frequency was higher during the first summer (68 % vs. 61 %) due to more Class I events (40 % vs. 27 %). Lesvos had an NPF frequency of 60 % during the summer 2021 campaign, with 40 % being Class I events. Thrace had a

frequency of 45 % during 2020 and 49 % during 2021, while the Class I events were 24 % and 20 %, respectively.

Sites with intermediate NPF frequency (between 20 % and 40 %) included Sifnos^{REM}, Finokalia^{REM}, and Helmos^{REM}. The frequency of NPF events at Sifnos was 33 % during 2020 and 36 % during 2021, with the Class I events accounting for 14 % and 16 %, respectively. A considerable difference in the event frequency between the two campaigns was observed in Finokalia, where NPF was 18 % (Class I, 7 %) and 35 % (Class I, 17 %), respectively. During 2021 the air in Finokalia was dryer (Tables S1 and S2) and the wind speed was higher than 2020. Those significant meteorological differences could explain the increased NPF frequency in Finokalia during 2021 compared to 2020. NPF events at Mt. Helmos occurred during 30 % of the summer days in 2020 and 27 % in 2021, with the Class I events only occurring on 12 % of all days in both campaigns.

Sites located mainly in the western and southern parts of Greece (Patras^{SUB}, Ioannina^{SUB}, NEO^{RUR}, Athens^{SUB}, and Chania^{URB}) were characterized by low NPF frequency (< 20 %). In Patras the event frequency was 15 % during 2020 and 10 % during 2021, while the Class I events were only three in total (Aktypis et al., 2023). The corresponding values for Ioannina were 17 % and 11 %, while the Class I events were also rare (only two in total). The NPF frequency in Athens was 14 % during 2020 and 9 % during 2021, while only three Class I events were observed exclusively during the first campaign. In Chania the frequency was 9 % and 3 % during the two campaigns, and all events were Class II with relatively low concentrations of formed particles. NEO (during summer 2020) had the lowest NPF frequency, with only two (4.5 %) Class II events occurring during the 45 d of the 2020 summer campaign. Nighttime bursts of nucleation mode particles that grew to larger sizes were occasionally

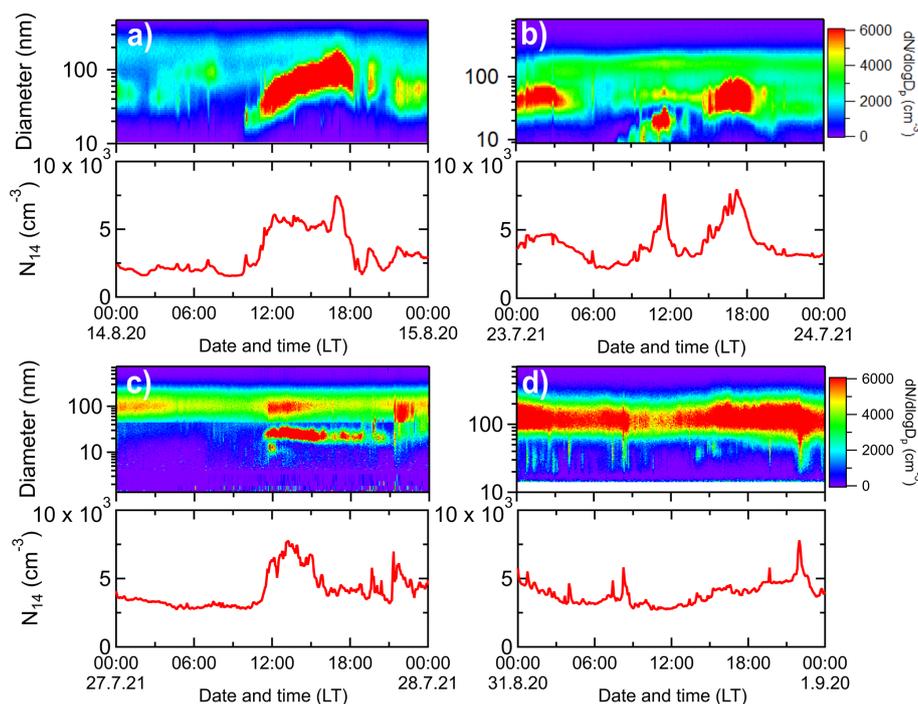


Figure 3. Typical examples of (a) a Class I NPF event occurring in Sifnos on 14 August 2020, (b) a Class II NPF event occurring in Finokalia on 23 July 2021, (c) an undefined event occurring in Patras on 27 July 2021, and (d) a non-event in Chania on 31 August 2020. The corresponding N_{14} during each day is also shown.

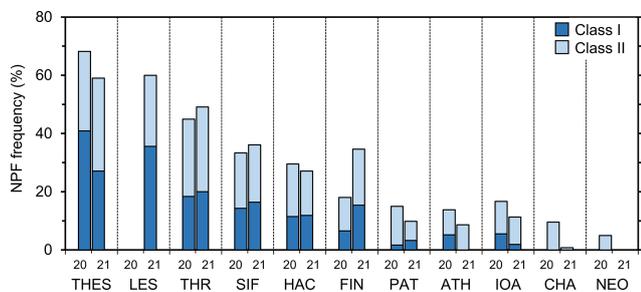


Figure 4. The frequency of Class I and Class II events observed at each measurement site for the summer campaigns of 2020 and 2021.

observed in NEO and will be analyzed in the next section. An example is depicted in Fig. S2, and these bursts were not counted as NPF events.

4.3 NPF event characteristics

In this section, various characteristics of the events like their possible relation with meteorological parameters, the condensation sink, and the growth rates are analyzed for each station. Meteorological data were compared, and the CS calculations were performed during the daytime and more specifically in the time window between 08:00–18:00 (LT), when NPF generally occurs. A t test was performed to com-

pare the CS, RH, and wind speed during the events and the non-events. The 99% confidence interval was chosen to reduce uncertainty in the comparisons. To evaluate the assumption that the data follow a normal distribution, the corresponding frequency and quantile–quantile (QQ) plots were analyzed. Excluding outliers, all distributions were close to normal.

At most sites, NPF took place during days with lower relative humidity. The average RH was significantly ($\alpha = 0.01$) lower during NPF days at six sites (CHA^{URB} , FIN^{REM} , LES^{REM} , THES^{URB} , THR^{RUR} , NEO^{RUR}) (Fig. S3). Lower RH during NPF has been observed worldwide (Kerminen et al., 2018) as well as in the Eastern Mediterranean (Kopanakis et al., 2013; Kalkavouras et al., 2020). There was no clear relation between wind speed and NPF events (Fig. S4). At five sites (PAT^{SUB} , FIN^{REM} , SIF^{REM} , THES^{URB} , THR^{RUR}) the wind speed was significantly higher ($\alpha = 0.01$) during NPF days, while the opposite was observed at two sites (ATH^{SUB} , CHA^{URB}). At the remaining sites, there was not a statistically significant difference. The corresponding ambient temperatures during events and non-events were also compared (Fig. S5). For most sites (THR^{RUR} , SIF^{REM} , FIN^{REM} , NEO^{RUR} , PAT^{SUB} , IOA^{SUB}) the temperature during event days was statistically the same compared to the non-event days. In THES^{URB} , LES^{REM} , and CHA^{URB} the temperature was on average a little higher (less than 1 °C) during NPF events, while the opposite was observed for HAC^{REM}

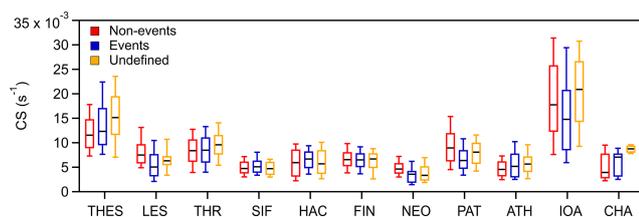


Figure 5. Comparison of the average condensation sink (CS) values in the time period between 08:00 and 18:00 LT during an NPF event, non-event, and undefined days at each station. The black lines represent the median, while the box edges are the 25th and 75th percentiles. The whiskers correspond to the 10th and 90th percentiles.

and ATH^{SUB}. This analysis suggests that temperature did not play a dominant role in determining the occurrence of NPF.

NPF events typically are associated with lower values of the condensation sink (Kerminen et al., 2018) because pre-existing aerosol absorbs the available condensable vapors quickly, preventing in this way the formation and growth of small clusters. The calculated condensation sinks were compared between the event, non-event, and undefined days for each location (Fig. 5). At most of the sites there was no considerable difference in the CS between events and non-events. This behavior has also been observed in Cyprus in the Eastern Mediterranean by Baalbaki et al. (2021). Only at 4 of 11 stations was there a significantly ($\alpha = 0.01$) lower CS during NPF events compared to the non-events. Three of these sites were in the western part of the country (Patras, NEO, and Ioannina), and the last one was Lesvos. Interestingly, in Thessaloniki, the numerous events were associated with a slightly higher CS than the non-events. Siakavaras et al. (2016) found that NPF in Thessaloniki occurs frequently during days with high levels of pre-existing aerosol. This may have to do with the origin of the air masses (analyzed in the next section) that arrive in Thessaloniki and can affect both NPF and PM concentrations. Overall, the intercomparison between the sites suggests that the average CS did not vary a lot at most locations and was relatively low during the measurement periods (around $6 \times 10^{-3} \text{ s}^{-1}$). This uniformity of the CS is consistent with the PM_{2.5} observations of Dimitriou et al. (2023) during the same periods. Thessaloniki and Ioannina were two exceptions with an average CS of 14×10^{-3} and $19 \times 10^{-3} \text{ s}^{-1}$, respectively. In Thessaloniki, the higher average CS can be explained by the central urban location of the site. The city of Ioannina was characterized by stagnation of the air (average wind speed was 1 m s^{-1}) during the study, but this alone cannot explain the high CS observed there. It is probably due to the combination of the low wind speed with the significant local sources that leads to the high average CS. This is not the case at other sites with comparable wind speeds like Thrace.

The average growth rate of all the sites was $5.7 \pm 2.9 \text{ nm h}^{-1}$ and varied from $3.4 \pm 1.8 \text{ nm h}^{-1}$ at the high-elevation site, Helmos, to $8.0 \pm 1.6 \text{ nm h}^{-1}$ in Fi-

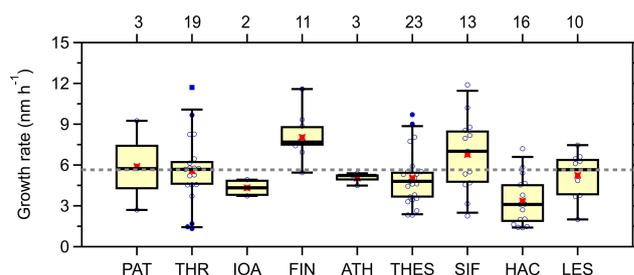


Figure 6. Average growth rates of the NPF events at each station (where Class I events were observed). The black lines represent the median, the red points the average, the box edges the 25th and 75th percentiles. The whiskers correspond to the 10th and 90th percentiles. The data points are shown with blue circles. The unfilled circles are data points that lie in between the 10%–90% range, while the filled ones are outliers. At the top, the number of data points is shown.

nokalia. Although there was a significant difference between these two locations, overall, the average growth rate at the remaining sites (Fig. 6) did not have a significant variability. The average growth rate observed is consistent with previous observations in the Eastern Mediterranean. In Helmos, the relatively low growth rates could be explained by the limited availability of condensable vapors in this high-elevation area (Sellegrì et al., 2019). On the other hand, the high growth rates observed in Finokalia imply high levels of vapors that condense rapidly. Relatively high growth rates in Finokalia during the warm period have also been reported in previous studies. Kalivitis et al. (2019) found that the 10-year average of the growth rate during the summer in Finokalia was equal to $7.3 \pm 3.9 \text{ nm h}^{-1}$. Even higher growth rates during summer have been reported by other relevant studies at this location (Pikridas et al., 2012; Berland et al., 2017; Kalkavouras et al., 2020). The potential linkages between the growth rate and meteorological conditions (T , RH, wind speed) for the sites that had enough growth rate data points (THES, THR, FIN, SIF, HAC, LES) were investigated. We did not observe any systematic dependency of the GR on any of these parameters. This suggests that other non-meteorological factors control the growth rate in these regions. The observed NPF frequencies and the growth rates are comparable with the ones reported for other areas in Europe during the summer by Nieminen et al. (2018). This also includes the small growth rates observed at the high-altitude sites which were also observed at Helmos.

4.4 Emissions of SO₂, NH₃, and biogenic volatile organic compounds in Greece during summer

Since the NPF frequency and intensity are largely dependent on the availability of the various gaseous precursors involved in nucleation (H₂SO₄, NH₃, volatile organic compounds, etc.; Kerminen et al., 2018; Lee et al., 2019), it is

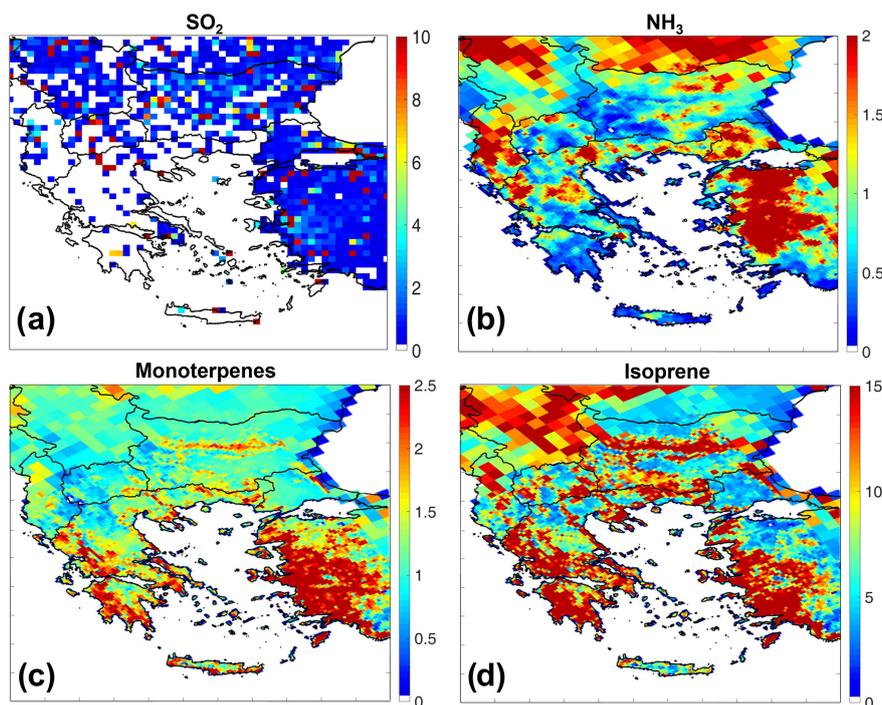


Figure 7. Emissions (in $\text{kg km}^{-2} \text{d}^{-1}$) of (a) SO_2 , (b) NH_3 , (c) monoterpenes, and (d) isoprene in Greece during the summer of 2020.

important to investigate their emission levels and spatial distribution across the country. Sulfur dioxide and ammonia emissions for the summer of 2020 were based on the CAMS-REG emission inventory (Kuenen et al., 2022) for the summer of 2018. We assume here that the emissions and especially their spatial distribution did not change drastically in this short period. Biogenic emissions were estimated by the Model of Emissions of Gases and Aerosols from Nature Version 3 (MEGAN3) (Guenther et al., 2012, 2020).

Sulfur dioxide (SO_2) levels can be a good indicator of sulfuric acid during summer in Greece. Significant amounts of SO_2 (Fig. 7a) are emitted in the Balkans (European Environment Agency, 2022) by coal or lignite-fired power plants. Although a significant reduction (almost 80 %) in SO_2 emissions has taken place in Europe between 2005 and 2020 (European Environment Agency, 2022), relatively high emissions are still present in the Balkans. The Maritsa Iztok complex in eastern Bulgaria for example is one of the largest SO_2 emitters in eastern Europe (Krotkov et al., 2016). Western Turkey also has elevated SO_2 emissions (Fig. 7a), due to the numerous power plants located there. Compared to these areas, Greece has lower SO_2 emissions. Elevated emissions in the Greek region are observed in the north, where the largest lignite-fired power plants of the country are located. Thus, higher SO_2 and therefore H_2SO_4 concentrations in the north are strongly associated with the highest NPF frequencies in the northern part of the country (i.e., THES^{URB} , THR^{RUR}). Although there are low SO_2 emissions in the southern and western parts of the country, these regions have adequate

sulfur levels during summer, due to both long-range transport from the north and also ship emissions (Dayan et al., 2017). The available SO_2 measurements in Patras revealed an average concentration of 0.42 ± 0.27 ppb that results in adequate levels of H_2SO_4 (in the order of 10^7 molec. cm^{-3}) for nucleation, at least in the western part of Greece. Sulfuric acid is assumed to be in pseudo-steady state in the gas phase with its production rate being proportional to the sulfur dioxide concentration and the OH radical concentration and inversely proportional to the condensational sink (Bardouki et al., 2003; Pikridas et al., 2012). We implicitly assume here that the reaction of sulfur dioxide with OH is the dominant source of sulfuric acid in the study area. The levels of OH were estimated using the predictions of the chemical transport model PMCAMx (Fountoukis et al., 2011) for the summer, while the sulfur dioxide and condensational sink were measured.

Significant spatial gradients in ammonia emissions are observed in Greece (Fig. 7b), dividing the country into two sub-regions. The central and northeastern parts are characterized by elevated emissions, exceeding $2 \text{ kg km}^{-2} \text{d}^{-1}$ in the agricultural areas of Thessalia, Thessaloniki, and Thrace. These are the largest agricultural areas of Greece, while livestock farming is also important there (Gemitzis et al., 2019; Hellenic Statistical Authority, 2021). Emissions of amines in these areas may also be elevated, since animal husbandry and agricultural activities are important amine and ammonia sources (Ge et al., 2011). The emissions in western and southern Greece are limited over smaller (mainly agricul-

4.5 Air mass origin during the NPF events

To gain insights into the potential nucleation mechanism(s) that dominate the Eastern Mediterranean, it is useful to investigate the air mass origin during the NPF events and compare it with the entire period and the non-event days (Wonaschütz et al., 2015). This analysis is restricted to the time window between sunrise and sunset (07:00–19:00 LT), when all the NPF events occurred.

4.5.1 Northeastern Greece

All three sites in northeastern Greece (THES^{URB}, THR^{RUR}, LES^{REM}) were dominated by northeastern air masses passing over the eastern Balkans and northwestern Turkey. Cluster analysis (Carslaw and Ropkins, 2012) was performed for the entire measurement period (Fig. 8a) to group all trajectories depending on their origin. An angle distance matrix was used, which, compared to the Euclidean, takes into account the movement patterns and shape of the trajectories more (Carslaw and Ropkins, 2012). The analysis revealed that during 44 % of all days, northeastern air masses prevailed in Thessaloniki. Air masses from the northeast also prevailed in Thrace (52 %) and Lesvos (60 %) during the whole period. Other important sectors included the northern and western Balkans and the south, where the corresponding trajectories passed over the Aegean Sea for at least 6 h before arriving at each site. Air masses from the east were common mainly in Thrace.

Most of the NPF events (43 %) in Thessaloniki were associated with air masses from the northern sector, while the corresponding fraction for the non-events was only 19 % (and for the whole period 27 %). These trajectories (Fig. S6) passed over North Macedonia, the Ptolemaida region (where the largest lignite-fired power plants of the country are located), and the agricultural area of Thessaloniki a couple of hours prior to arriving at site. The increased potential of these air masses to form new particles can be explained by the fact that they are expected to be rich in both SO₂ and NH₃ due to the particular route they follow (Fig. 7a, b). A large fraction (38 %) of the events was associated with trajectories from the northeast, usually passing over eastern Bulgaria or northwestern Turkey 10–20 h before arrival. These air masses also spent some time over the agricultural region of Thrace, located about 160 km to the east of the site. Air masses originating from the southwest were moderately associated with NPF in Thessaloniki.

Northeastern winds (Fig. S7) passing over eastern Bulgaria were prevailing in Thrace during 57 % of the NPF events, while the corresponding fraction for non-events was 33 %. NPF under eastern air masses was also frequent in Thrace (34 %). There was not a single event associated with southern wind directions, a condition present during 15 % of the days.

In Lesvos almost all NPF events took place when the air masses were northeastern (Fig. S8), passing over western Turkey or eastern Bulgaria. Only two events occurred when air masses came from the south. Being downwind of the northwestern Turkey region, the corresponding air masses are expected to be rich in SO₂ since several lignite-fired power plants are located there (Say, 2006; Vardar and Yumurtaci, 2010). During all five NPF events in Lesvos under northern air masses, NPF was also observed in Thrace on the same day. This suggests that these occurred on a spatial scale of more than 200 km.

4.5.2 South Aegean

The sites located in the south Aegean (SIF^{REM}, CHA^{URB}, and FIN^{REM}) were exposed to the northeastern flow of the Etesians. During 44 % of the measurement days, northeastern air masses were reaching Sifnos, while the corresponding fractions were 48 % for Finokalia and 69 % for Chania (Fig. 8b). Northern flows from the Balkans and western air masses mostly of marine origin were also observed frequently (25 %–50 %).

Most of the NPF events in Sifnos (73 %) occurred in fast-moving air masses from the northeast (Fig. S9). Only three events were associated with trajectories from the west that in all cases had spent several hours over the Peloponnese. Not a single event occurred in southwestern air masses, which were common during non-events. During the summer of 2021, 15 of 16 events that occurred in Sifnos also occurred in Lesvos (280 km upwind). Considering that these events typically lasted for more than 4 h, they appear to take place on a regional scale. However, additional measurements in the Aegean are needed to confirm that this is indeed happening. Kalkavouras et al. (2017), analyzing NPF events in Santorini, a nearby island, during a short-term summer campaign (15 to 28 July 2013), found that under the Etesian flow, their spatial extent can be at least 250 km. However, the fact that the NPF frequency in Sifnos is almost half than that in Lesvos suggests that as the air masses travel over the sea, the concentrations of the gaseous precursors become progressively lower, resulting in fewer events.

All the three weak NPF events in Chania occurred when the air was coming from the north (Fig. S10). In all cases the trajectories passed over land areas of the eastern Evia and Attica regions about 10 h before arriving at the site. Chania was also exposed to the northeastern flow that appears to favor NPF in Sifnos and Lesvos, but a very low NPF frequency and weak events were observed under these conditions.

The majority (69 %) of the events in Finokalia were also connected with northern and northeastern air masses (Fig. S11). Of the 16 events occurring under northeastern flow, 11 also took place in Sifnos and Lesvos, suggesting regional NPF. An important fraction (31 %) of the events in Finokalia occurred when the wind was coming from the west. The air masses in these cases had spent 2–6 h over

mainland Crete before arriving at Finokalia, unlike Chania (145 km west), where they arrived directly at site after spending > 12 h over sea. These observations (including the five remaining events with northeastern air masses that occurred only in Finokalia) suggest that NPF events on a relatively small spatial scale (less than 150 km) are possible in this area. These “local” events were estimated to account for 36 % of the total. An example of such an event is shown in Fig. S12. Our explanation regarding these local events in Finokalia is that the air masses that both come from the northeast shift and pass over Crete (Kalkavouras et al., 2021; Rizos et al., 2022) picking up ammonia that is emitted in the agricultural area of central Crete.

4.5.3 Western and central Greece

The sites in the western and central parts of the country (PAT^{SUB}, NEO^{RUR}, IOA^{SUB}, and ATH^{SUB}) all had an NPF frequency of less than 20 %. During the measurement period, clean air masses from the western sector (Fig. 8c), typically spending more than 12 h over the Ionian Sea, arrived at the western sites (IOA^{SUB}, PAT^{SUB}, and NEO^{RUR}). In Athens air masses from the northeast prevailed (during 45 % of the days), while northern, western, and southern directions were also observed.

In Patras during the two strongest Class I events, air masses passed over the agricultural region of Iliia, south of Patras. In total, 31 % of the days were classified as transported events (Aktypis et al., 2023) with the estimated region of NPF being an agricultural area 150 km northeast of Patras.

Two-thirds of the NPF events (including all three Class I events) occurred in Athens when the air masses were coming from the west or the northeast (Fig. S14). In all cases, they passed over the greater Boeotia region (which has higher NH₃ emissions due to agricultural activity). It is worth mentioning that, excluding the NPF events, the western sector was not a frequently traversed pathway for the air masses (Fig. 8c). The other 33 % were weak Class II events which occurred when the wind was coming from the north–northeast and passed over central Evia, where NPF could also be happening.

In Ioannina, NPF was favored by air masses passing from North Macedonia and Albania (Fig. S15). The only two Class I events observed in Ioannina occurred in air masses originating from North Macedonia and also passing over the Ptolemaida region and the agricultural area of Thessaloniki. Air masses from land areas of Albania were also associated with (weaker) events. Although Ioannina is in close proximity to the various sources of SO₂ and NH₃, the mountain range of Pindus, which separates western from eastern Greece, limits the passage of the air masses from the northeast. The lack of gaseous precursors together with the high condensation sink observed there and the meteorological conditions (high RH, air stagnation) could be part of the explanation why NPF is infrequent in this area.

Two of the three NPF events observed in NEO occurred in northeastern air masses, passing over the Peloponnese before arriving at the site. In NEO, there were at least three cases where nucleation mode particles appeared during afternoon and nighttime hours and also showed signs of growth to larger sizes (Fig. S2). The trajectory analysis for these days revealed that these particles could be a result of transport from the Iliia agricultural region, located 100 km north. This region has also been found to affect NPF in Patras, when the air passes over it (Aktypis et al., 2023).

4.5.4 High-elevation site

The air masses during 70 % of the NPF events in Helmos were northeastern, while the corresponding fraction for the whole measurement period was 49 %. The remaining 30 % of the events occurred when the air came either from the southwest or the northwest. The NE air masses passed over the greater agricultural area of Boeotia a few hours before arrival. This is the same area that appears to affect the nearby stations Patras and Athens. There were also many cases (about 45 % of all days) where distinct particle modes in the nucleation and Aitken range appeared during the early afternoon (around 14:00 LT). These modes did not grow and were probably the result of transported events (traditionally classified as undefined). This lack of further growth is probably due to the limited levels of condensable vapors in this high-elevation area.

4.6 Concurrent events at multiple sites

There was not a single day when NPF events were observed at all stations. This is reasonable considering the topographic heterogeneity of Greece and that each station was exposed to different air masses. NPF events took place at most (50 %–85 %) of the sites during 10 d. During 7 of the 10 cases, the predominant wind direction was from the northeast. Two cases occurred when the air masses originated from the west (Lesvos and Thrace continued to be affected by eastern air masses in one case and also had NPF). The last case occurred when the air masses originated from the north (passing over North Macedonia and Bulgaria) at all stations. Although the condensation sink in most cases was lower than the corresponding average of each station for the whole period, it was not statistically significant ($\alpha = 0.01$). It was significantly lower by 50 % only during the two cases when the air masses were arriving from the west, spending many hours over the Ionian Sea. Being primarily of marine origin, these air masses were relatively clean but appeared to have enough gaseous precursors in order for new particles to be formed. This can be explained by the fact that in all cases they spent some hours over land areas before arriving at each site and did not come directly from the sea.

The rarity of simultaneous events occurring at over half of the sites during the measurement period highlights the di-

verse nature of the atmosphere across the different locations in terms of its availability in NPF precursors.

5 Conclusions

Two summers (2020 and 2021) of extensive continuous particle number size distribution measurements at 11 different locations in Greece were analyzed to quantify the NPF frequency and its spatial extent across the country and also assess the factors affecting them.

A significant variability in the NPF frequency was observed among the different sites, varying from close to zero in the southwestern parts of the country to more than 60% in the northern central and eastern regions. An average particle growth rate of $5.7 \pm 2.9 \text{ nm h}^{-1}$ was calculated for all sites and varied from 3.4 nm h^{-1} at Mt. Helmos to 8 nm h^{-1} in Finokalia. The average CS of events and non-events was similar at most of the measurement sites (6 out of 11).

The southern and western parts of the country, where NPF was infrequent, were characterized by lower and more localized ammonia emissions. Although the emissions of biogenic volatile organic compounds were higher in these areas, they did not appear to lead to enhanced new particle formation. The detailed analysis of the air mass trajectories revealed that the infrequent clear NPF events at these sites occurred when the air masses had spent a few hours over land areas with agricultural activities and thus elevated ammonia emissions. Air masses arriving directly from the sea were not connected with NPF at all sites. The high-NPF-frequency sites in the north were in close proximity to large agricultural areas with the highest ammonia emissions of the country (higher than $2 \text{ kg m}^{-2} \text{ d}^{-1}$). The corresponding air masses during the NPF events in these areas were also enriched with emissions from various big coal-fired power plants located in northern Greece and the neighboring countries. Many regional events were observed at the sites of Lesvos, Sifnos, and Finokalia, all located in the Aegean Sea. However, in Finokalia there were also events (36%) that were not observed at the nearby stations, making them more localized ($< 150 \text{ km}$). This supports the hypothesis that a compound is missing (probably ammonia) from those air masses, which they collect after passing over central Crete, increasing in this way their potential to form new particles on a smaller spatial scale.

There was not a single day where NPF events took place at all the measurement sites. Simultaneous events occurred at at least half of the sites on only 10 of 124 d. In most of the cases, northeastern air masses arrived at the sites. The fact that concurrent events were extremely rare throughout the measurement period signifies the heterogeneity of the atmosphere at the different locations regarding the availability of NPF precursors.

Data availability. The field measurements are available from the authors (spyros@chemeng.upatras.gr).

Supplement. The supplement related to this article is available online at: <https://doi.org/10.5194/acp-24-65-2024-supplement>.

Author contributions. AA was responsible for the summer 2021 measurements in Patras, Xanthi, and Ioannina and analyzed the data and wrote the paper. DP performed the stimulations and calculated the emissions of SO_2 , NH_3 , and organics. CK, AM, CNV, and KF helped with the measurements in Patras. EK helped with the measurements in Thrace. PK, AB, and NM were responsible for the measurements in Lesvos and Sifnos. NK was responsible for the measurements in Finokalia. KE, SV, and MIG were responsible for the measurements in Athens and Mt. Helmos. AK and CS were responsible for the measurements in Thessaloniki. ML and SEC were responsible for the measurements in Chania, and SNP was responsible for the design of the study and the synthesis of the results and contributed to the writing of the paper.

Competing interests. The contact author has declared that none of the authors has any competing interests.

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